



CASE SU-7152

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

HASSAN Y. ELNAGAR, ET AL.

APPN. NO.: 09/484,844

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PROCESS FOR PRODUCING
N-HALOGENATED ORGANIC
COMPOUNDS

GROUP ART UNIT: 1626

EXAMINER: LAURA STOCKTON

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

DECLARATION UNDER 37 CFR 1.312

David Shelton hereby declares as follows:

I am a research supervisor employed by Albemarle Corporation, the Assignee of the above entitled application by unrecorded Assignment. In 1973 I received a B.S. degree in pharmacy from the University of Cincinnati and in 1974 I received a M.S. degree in pharmaceuticals from the University of Cincinnati. I have been continuously employed by Albemarle Corporation in a capacity of research supervisor since February 1998. I supervised the synthesis work performed at Albemarle Corporation, and coordinated with the independent laboratory for the testing described herein to determine the dusting properties of the samples.

The synthesis work to prepare the samples for which the test results are reported herein was performed by Jeffrey Todd Aplin at the research laboratories of Albemarle Corporation in

Baton Rouge, Louisiana, as described in his Declaration already on file in this case. The described dusting tests were conducted by EMSL Analytical, Inc., an independent testing laboratory in Westmont, New Jersey.

The work that EMSL Analytical, Inc. was requested to perform was to conduct dusting tests upon 1,3-dibromo-5,5-dimethylhydantoin samples from

- (1) Example 10 in Column 5 of Waugh et al. U.S. Pat. No. 3,121,715;
- (2) Example III in Column 4 of Cole U.S. Pat. No. 4,621,096;
- (3) Example III in Column 4 of Cole U.S. Pat. No. 4,621,096, with the modification of a water wash following the calcium hydroxide treatment; and
- (4) Albemarle product made in accordance with a process of the present application.

Methodology

The test used by EMSL Analytical, Inc. was an elutriation test, in particular, modified EPA method # 540-R-97-028, "Superfund Method for the Determination of Releasable Asbestos in Soil and Bulk Materials," in which the rate of release of respirable dust is evaluated from measurements of the mass of the dust collected over time on the set of filters mounted over the opening of the elutriator.

The analysis was performed using an elutriator (dust generator) per the specifications provided in EPA 540-R-97-028. Auxiliary equipment used to support the dust generator included a 129 hp DC motor to drive the tumbler, a vacuum pump capable of drawing 20 L/min at minimum load (run at 1 to 2 L/min), and variable flowmeters.

Prior to using the dust generator, a supply of 0.45 μm pore size, 25 mm diameter mixed cellulose ester (MCE) filters must be conditioned for at least 24 hours at constant humidity and temperature. After conditioning, the filters are pre-weighed. The sample to be used in the dust generator should be dry. If the sample is visibly wet, it should be dried at low temperature or stored for several days in a desiccator. Once dry, a sample weighing between 50-80 g is loaded into the tumbler. Prior to initiating a run, the sample is conditioned in the tumbler with no tumbling and with air flowing through the system for at least four hours. Once the sample has been conditioned, the tumbler is set to rotate at selected rotation speed and the rate of respirable dust generation is monitored by recording the weights of a set of filters that are sequentially changed out of the filter mounts. The ideal weight of dust to be deposited on each filter is between 0.01-0.03 g because a higher loading would increase the risk that a portion of the deposit on the filter drops.

Elutriation experiments for each sample were conducted under the following conditions: approximately 50g of starting material, a tumbler rotation rate of 60 rotations/min, 4 L/min flow rate of air through the elutriator system, ~45% relative humidity, and a total collection time of 70 minutes. The filter cassettes were removed from the mounts on the top of the column at predetermined times to measure the weight of collected dust. The samples were tested as received.

An additional elutriation experiment was conducted on the sample from Example 10 in Column 5 of Waugh et al. U.S. Pat. No. 3,121,715 to explore its behavior at low humidity. For this additional experiment, the sample was dried overnight in a vacuum desiccator prior to the

experiment. Conditions also differed from the 45% relative humidity experiments in that air was passed through a desiccant chamber before entering the tumbler and a higher flow rate was used.

Calculations

The cumulative mass collected on the filters is plotted as a function of time. The cumulative mass released from the sample over time is calculated using the relationship:

$$M_s = (F_s + F_d + F_c)/F_c * M_f$$

where

M_s is the cumulative mass of dust released from the sample between the start of the run at time “t” (g)

M_f is the cumulative mass collected on filters between the start of the run at time “t” (g)

F_s is the rate of airflow through the scrubber (cm^3/g), and

F_d and F_c are the rates of airflow through the opening of the elutriator.

The total mass of dust in the sample at the beginning of the run is estimated using:

$$\ln (M_o - M_s) = \ln M_o - kt$$

where

M_o is the mass of dust in the sample at the beginning of the run (g)

k is the first-order rate constant for the release of dust from sample (s^{-1}), and

t is the time since the start of the run (s).

A plot of $\ln (M_o - M_s)$ versus t produces a slope equal to the rate constant for the release of dust from the sample. The value of M_o and k are determined by regression fit.

The mass percent of respirable dust in the sample is then calculated as follows:

$$\%RD = 100 * M_o/M_{\text{sample}}$$

where

%RD is the mass percent of respirable dust in the sample (%)

M_o is the mass of dust in the sample at the beginning of the run (g), and

M_{sample} is the mass of the original sample placed in the tumbler (g).

Results

The data are plotted in attached Figure 1. In the Figure, sample 318363 was made according to Example III in Column 4 of Cole U.S. Pat. No. 4,621,096; sample 318364 was made according to Example III in Column 4 of Cole U.S. Pat. No. 4,621,096, with the modification of a water wash following the calcium hydroxide treatment; sample 318362 was made according to Example 10 in Column 5 of Waugh et al. U.S. Pat. No. 3,121,715; and sample 329085 is Albemarle product made in accordance with a process of the present application.

The rate of release of respirable dust for the samples tested near 45% relative humidity was lowest for the Albemarle product made according to the presently claimed invention. The sample treated with $\text{Ca}(\text{OH})_2$ without a water wash had more dust than did the sample made according to the present invention, and the sample treated with $\text{Ca}(\text{OH})_2$ followed by a water wash released more respirable dust than did the $\text{Ca}(\text{OH})_2$ -treated sample without the water wash.

For the sodium carbonate treated sample, no measurable weight was collected for the 45% relative humidity elutriation experiment. Upon completion of the experiment, a significant

fraction of the material was found adhered to the walls of the tumbler. Remaining material was found agglomerated in globules. By comparison, the other three samples showed a much lower tendency for agglomeration and adhesion to the walls of the tumbler. The low relative humidity experiment on this sodium carbonate treated sample had no adherence to the tumbler walls, and produced considerable dust, more than was produced by the sample treated with $\text{Ca}(\text{OH})_2$ followed by a water wash.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the above-identified application when it issues as a patent.

12/16/03

Date



David Shelton



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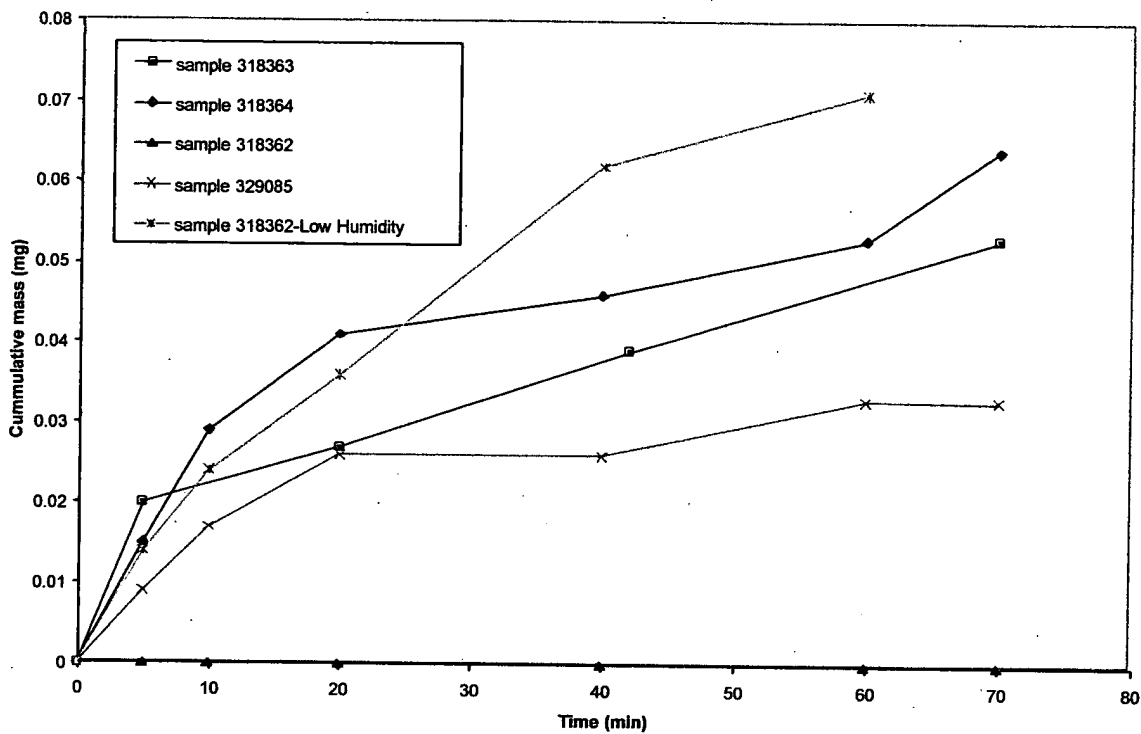


Figure 1. The cumulative mass of the material collected on the filters versus collection time.